Superconducting quantum interference device magnetometry during ultrahigh vacuum growth

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An *in situ* study of the absolute magnetic moment of thin ferromagnetic films grown in ultrahigh vacuum (UHV) is described. The moments are measured using a newly innovated technique, rotating sample superconducting quantum interference device (SQUID) magnetometry (RSSM), in which the sample is spun at low frequency close to an *ex situ* second derivative superconducting pickup loop structure coupled to a rf SQUID. The sensitivity of the measurements is $\approx 10^{-6}$ emu, which corresponds to 1/10 of an atomic layer of Fe over a 0.5 cm^2 area, and the geometry of the pickup loops makes the magnetometer very insensitive to background laboratory noise. The moment of Cr grown on Fe was measured. It was determined that the first two layers of Cr have an average moment of 0.75 μ_B per atom, and are antiferromagnetically aligned to the Fe film moment. The simplicity of RSSM and the fact that the absolute moment is measured makes this a powerful new approach for characterization of magnetic samples in UHV.

The past decade has seen a resurgence of interest in magnetic materials, with a special emphasis on ultrathin films, magnetic surfaces, and interfaces.[1] Although new fabrication techniques have been developed to synthesize these materials and new tools of surface science have been exploited to monitor the structure and chemistry, until very recently[2] there have been no reports of concomitant development in techniques to obtain absolute measurements of the magnetic moment in a typical ultrahigh vacuum (UHV) deposition system. The popularity of the magneto-optical Kerr effect to monitor magnetization of films in UHV attests to the need for such measurements, but optical measurements are very difficult to calibrate for absolute values. Absolute magnetometry is of course the most fundamental measurement to make on a magnetic material. It is usually measured *ex situ* after the film is covered with a nonmagnetic overlayer.[3–5] For ultrathin films, i.e., 1–10 atomic layers (AL), this method is unwieldy because many samples must be made in order to characterize the moment as a function of thickness, inducing a variety of systematic errors into the measurement. In addition, interesting interfacial effects such as the predicted enhancement of the moments due to reduced coordination, the effects of intermixing, and induced moments on nonmagnetic adatoms are lost by this method. Magnetometers using induction coils which can be applied to samples in UHV are typically constrained to having the sample mounted on a quartz rod, thus making high temperature growth and annealing studies difficult.[6] Many magnetic thin-film systems also must be grown on lattice-matched metallic substrates, which are not compatible with the induction measurement. For these reasons, we have developed a rotating sample superconducting quantum interference device (SQUID) magnetometer (RSSM) which can be applied to a sample inside an UHV chamber. The high sensitivity of SQUIDs to changing magnetic fields at low frequency allows us to mechanically spin the sample at 8 Hz and isolate the signal from very thin ferromagnetic films grown on any substrate. We have also found that we are able to run the SQUID unshielded in a laboratory environment in close proximity to a combination of typical laboratory equipment.

In this letter we demonstrate an *in situ* measurement of the moments of ultrathin Fe films using RSSM with a sensitivity of 10^{-6} emu. This sensitivity corresponds to the moment of $\approx$0.1 AL of Fe (1 AL=1.44 Å) over a 0.5 cm^2 area. The films were grown in an UHV chamber, and the magnetometer consisted of a rf SQUID and accompanying second derivative pickup loop structure which were housed in a liquid helium dewer.[7] Although this *in situ* magnetic characterization is applicable to a variety of investigations, it is especially valuable for a study of the moment versus film thickness because the measurements can be performed on the same sample as the thickness is increased. This eliminates the need for making a large set of individual samples, removing them from the system, and measuring their moments separately on another apparatus, a procedure which is not only time consuming, but raises questions of reproducibility and reliability.

Fe grown on GaAs(001) was selected as a test system for this study due to its low coercivity and square hysteresis loops. In the chamber, shown in Fig. 1, a polished and chemically etched single-crystal GaAs(001) substrate was mounted on an x-y-z sample manipulator with a differentially pumped rotatable feedthrough. The substrate was oriented in such a manner that the Fe film grown would have one of its magnetic easy axes perpendicular to the rotation axis. The sample manipulator was designed so that the sample could be moved from the growth position to a magnetizing coil, and then under the second derivative pickup coil of a rf SQUID where it was spun at a frequency of 8 Hz.

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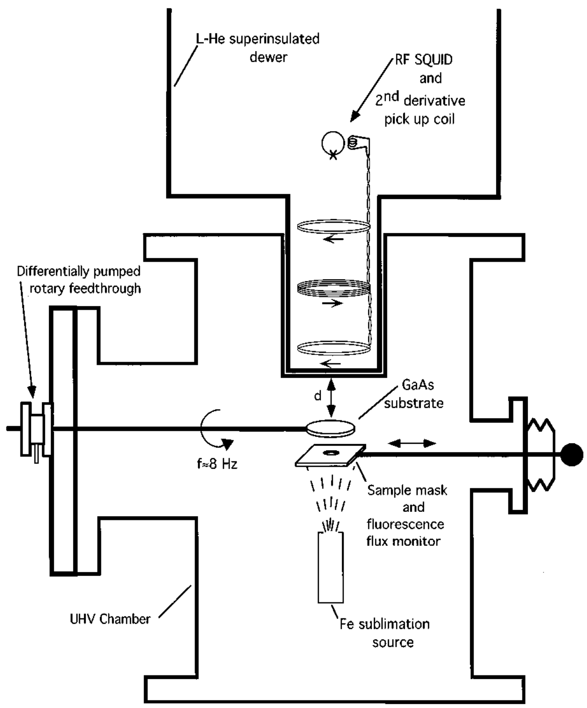

FIG. 1. Schematic of the UHV chamber used for the *in situ* SQUID magnetometry of ultrathin films.

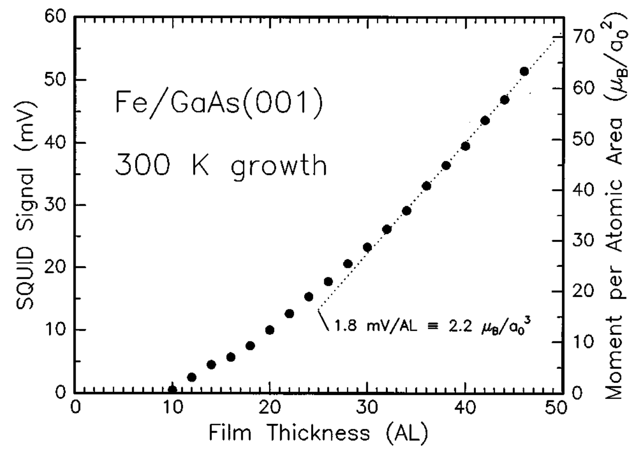

FIG. 2. The SQUID signal from an Fe film on GaAs as the thickness is increased from 0 to 48 AL. The asymptotic slope is shown as a dotted line in the figure, and corresponds to a SQUID signal of 1.8 mV/AL on the 0.5 cm^2 Fe film as the thickness is increased. The scale on the right-hand side was calculated using the bulk Fe magnetization of 2.2 μ_B/atom for the asymptotic slope.

The rotation of the sample was achieved using a flow regulated air turbine motor, which was used instead of an electrical motor in order to minimize the magnetic signal from the driving motor. Before use, the turbine was degaussed, and during use it was housed inside a mu-metal can. An Fe film was deposited on the substrate in fixed increments, and the 8 Hz signal from the SQUID was measured after each increment. During the depositions, a 0.5 cm^2 aperture mask defined the area of the film and prevented deposition onto the sample support shaft. Flux falling upon the substrate first passed through the sensor box of an Inficon Sentinel Rate Monitor, which measures the fluorescence of atoms passing through the box. Prior to use, the Sentinel was calibrated against both a quartz crystal microbalance and a Kevex x-ray fluorescence monitor.

The SQUID signal at the 8 Hz rotation frequency was monitored with a two-phase lock-in amplifier, that recorded both the magnitude and the phase of the signal relative to the angle of the spinning sample (which was monitored with a dynamically balanced light chopper). After the GaAs substrate was heated to 600 °C to desorb oxygen and prepare the surface, an Fe film was deposited at 300 °C. Results for a representative film grown in 2 AL increments are shown in Fig. 2. It can be seen that there is no remanent magnetic moment in the film up to about 10 AL, which is interpreted as a quenching of the Fe moment at the interface.[3] As the thickness is increased, the curve is slightly concave, indicating that there may be some additional quenching due to, e. g., Ga or As intermixing, occurring up to 30 AL. After about 30 AL of Fe was deposited, the asymptotic slope of the SQUID signal was $\approx 1.8 \pm 0.1$ mV/AL of Fe. For confirmation that the signal was coming from the Fe film, the sample magnetization was flipped into the opposite direction by passing a current pulse through the nearby magnetizing coil. It was observed that the phase of the signal shifted by 180° after the magnetization reversal. This change in phase proves that the signal is coming from the deposited Fe, since the same test had been conducted on the bare substrate prior to deposition with a null result. We note that this data, acquired in approximately 3 h, is equivalent to measuring 20 individual epitaxially grown samples. At the typical rate of one/day, this would amount to four weeks of sample preparation alone. To this must be added the time needed to perform magnetometry measurements on these 20 samples.

An absolute calibration of the system was conducted by mounting a ten turn, 8-mm-diam coil in the sample position. A 120 μA (zero to peak), 8 Hz sinusoidal current was passed through the calibration coil, which generated a signal equivalent to that of a 10-Å-thick Fe film of the same 0.5 cm^2 area (assuming a moment of 2.2 μ_B/atom). The signal from the calibration test coil was measured as a function of the distance from the center of the coil to the bottom of the stainless steel sleeve into which the dewer was inserted. These data are shown in Fig. 3. The magnitude of the signal as a function of distance agrees within 10% of the calculated response of the magnetometer. However, at the sample position of $\approx$13 mm from the bottom of the stainless steel dewer sleeve a signal of 1.3 mV/AL was measured compared with the 1.8 mV/AL shown in Fig. 2. This discrepancy is largely accounted for by the fact that the shaft wobbles by several millimeters when it is spinning, and there is also a slight uncertainty in the area of the calibration loop. Due to these types of systematic errors, we have calibrated the magnetometer using the bulk Fe magnetization as an internal standard when making measurements. The frequency response of the magnetometer while mounted in the stainless steel sleeve was also checked, and was flat out to at least 1 kHz. Additional tests showed that the presence of the stainless steel dewer sleeve does not reduce the amplitude of the signal.

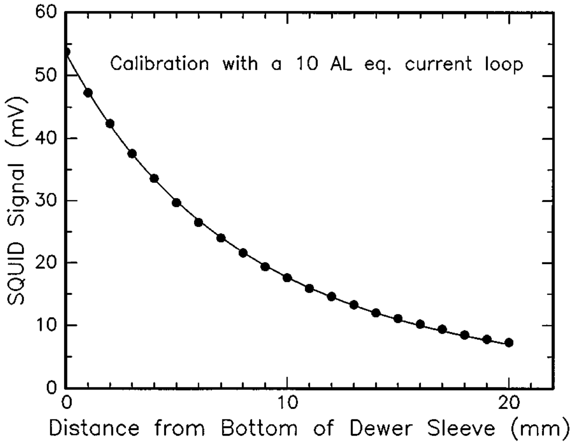

FIG. 3. The SQUID signal from a calibration coil (equivalent to 10 AL of Fe deposited over 0.5 cm^2) as the distance from it to the bottom of the dewer is changed. The solid line shows a calculated fit assuming the coil is a rotating magnetic dipole with its axis of rotation perpendicular to the axis of the pickup loops.

As a further demonstration of the utility of RSSM we have measured the decrease of the moment of an Fe film as additional layers of Cr are deposited. These data are shown in Fig. 4, where it can be seen that the first two layers of Cr decrease the total moment of the film by 1.5 μ_B/a_0^2, where a_0 is the lattice constant of bulk bcc-Fe. This is due to the antiferromagnetic coupling of the first two Cr layers to the Fe moments,[8] and can be interpreted as an average of $\approx$0.75 μ_B magnetization on each Cr atom in the first two layers. These data show a much smaller Cr moment than the results of Ref. 2, however, our result is more in line with that estimated using well-accepted sum rules applied to core-level photoemission and x-ray magnetic circular dichroism.[9]

The noise in the SQUID magnetometer system at 8 Hz was on the order of 10^{-2} $\phi_0/\sqrt{\mathrm{Hz}}$, where ϕ_0 is the magnetic flux quantum. This noise level corresponds to a field noise of 10^{-7} G/$\sqrt{\mathrm{Hz}}$ at the lowest pickup loop of the second derivative gradiometer, and is about 100 times that of the system when it is in a mu-metal shielded environment. Therefore, the noise in the measurements is totally dominated by the laboratory magnetic background originating at nearby cryopumps, roughing pumps, and other electrical equipment. The impact of this noise is greatly reduced by the use of the second derivative pickup loop configuration, even though the tail of the dewer is slid into a metal sleeve in the vacuum chamber. The noise goes down only by about a factor of 2 when the dewer is removed from the sleeve and placed in a nonferrous stand a few feet from the UHV system. Therefore, with the present SQUID system there is opportunity for considerable improvement in sensitivity with more sophisticated shielding. It is also interesting to note that the base field noise level is well above that already demonstrated with high-T_c SQUIDs, and on the order of that achieved by a state-of-the-art flux gate magnetometer. It should, in fact, be feasible to operate a high-T_c SQUID *in situ*, which would result in lower noise due to a more compact geometry and a higher signal due to being closer to the sample. Either a high-T_c SQUID or a flux gate implementation would require a gradiometer configuration to reduce the laboratory background noise.

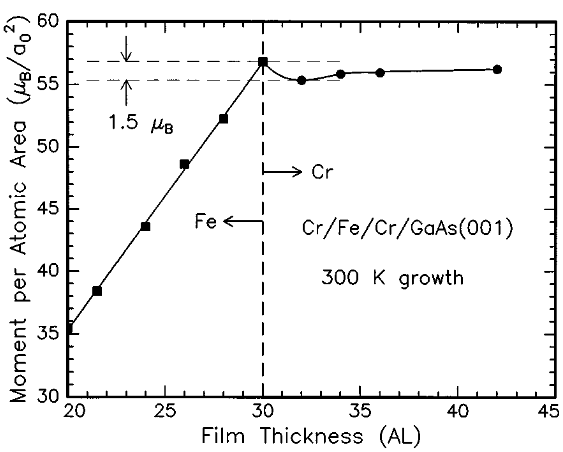

FIG. 4. Total remanent moment as Cr is deposited on top of an Fe film. A buffer layer of 5 AL of Cr was deposited on the GaAs in order to reduce interdiffusion. The scale on the vertical axis is determined using the bulk magnetization of Fe for the slope of the signal up to 30 AL.

This study demonstrates the feasibility of the *in situ* measurement of the total moment of ultrathin films using a SQUID magnetometer. It also represents the first *in situ* SQUID-based measurement of the absolute moment versus thickness of Fe and Cr layered structures. This technique is relatively simple and robust, and should be capable of providing useful information on a number of other ferromagnetic thin-film systems. In this study we have resolved 0.1 AL moments, and we expect to be able to achieve much higher sensitivity by improving the mechanical stability, magnetic shielding, and ultimately by using an *in situ*, high-T_c-based magnetometer.

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